

Application No. 10/058,707
Reply to Office Action of July 6, 2005

in view of JP '852 and U.S. Patent Application Publication No. 2001/0018162 of Kida ("Kida") or U.S. Patent No. 6,090,506 of Inoue et al. ("Inoue"). Finally, claim 15 has been rejected under 35 U.S.C. § 103(a) as being unpatentable over Takami in view of WO '228 and further in view of JP '852 and U.S. Patent Application Publication No. 2002/0001756 of Hamamoto ("Hamamoto"). Applicants respectfully traverse these rejections and the arguments in support thereof for the reasons set forth previously on the record, which Applicants rely upon in full, and for the additional reasons which follow, and respectfully request reconsideration and withdrawal of the rejections.

The Present Invention

As previously explained on the record, the present invention is directed to a non-aqueous electrolyte secondary battery which contains a non-aqueous solvent containing: (A) a cyclic carboxylic acid ester; 0.5 to 20 volume % of (B) a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond; and (C) a cyclic carbonic acid ester having no carbon-carbon unsaturated bond. Component (B) comprises vinylethylene carbonate (VEC) and vinylene carbonate (VC). As previously explained and shown, the combination of VC and VEC is particularly desirable because VEC suppresses the reaction activity of VC at high temperatures to improve storage characteristics of the battery at such temperatures. Further, VEC is an important component of the non-aqueous electrolyte which is decomposed by reduction faster than the solvent at a slightly more noble potential than 0 V (Li/Li⁺). Since the unsaturated bond is prone to polymerization, VEC undergoes a chain reaction on the negative electrode and rapidly forms a close and strong film on the negative electrode surface. This film serves as a physical barrier which inhibits the contact of solvent molecules around lithium ions with the negative electrode, thereby suppressing the reductive decomposition of the cyclic carboxylic acid ester on the negative electrode (see page 5, lines 18-25). As previously explained and demonstrated, the presence of LiBF₄ and LiPF₆ in a molar ratio of 1:9 to 9:1 in the solute (as recited in claim 14) improves cycle characteristics of the battery and decreases the amount of gas generated, and also results in favorable capacity maintenance rate and cycle life.

Double-Patenting Rejection Based on the '473 Patent in view of Takami

Regarding claims 1-7, the Examiner argues that the '473 patent discloses a non-aqueous electrolyte secondary battery, but acknowledges that the use of a cyclic carbonic acid ester having no carbon-carbon unsaturated bond is not taught. However, Takami allegedly discloses a non-aqueous electrolyte secondary battery comprising an electrode group containing a positive electrode, a negative electrode and a non-aqueous electrolyte including solvents and lithium salts dissolved therein. It is allegedly disclosed to use cyclic carbonates, such as ethylene carbonate (EC), propylene carbonate (PC), or vinylene carbonate (VC), together with γ -butyrolactone (GBL), and further to be desirable to prepare a mixed solvent including an aromatic compound. Various preferred combinations of non-aqueous solvents are allegedly taught, particularly in paragraphs [0061] and [0174] - [0176].

Therefore, the Examiner concludes that it would have been obvious to one having skill in the art at the time of the invention to use the cyclic carbonic acid ester having no carbon-carbon unsaturated bond of Takami in the electrolyte solution of the battery of the '473 patent because Takami teaches that it is desirable to use such a cyclic carbonic acid ester solvent in combination with other known solvents to improve the charge-discharge efficiency and cycle characteristics. Additionally, Takami allegedly teaches preferred combinations including GBL, EC and VC; GBL, PC and VC, and GBL, EC, PC and VC. Applicants respectfully traverse this rejection as follows.

The non-aqueous solvent of the '473 patent contains a cyclic carboxylic acid ester and a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond. Although exemplary cyclic carbonic acid esters include VC and VEC, the combination of VC and VEC is not described nor included in any of the Examples. Takami teaches a mixed non-aqueous solvent which contains (GBL) as a main component, combined with a cyclic carbonate such as (PC), (EC), or (VC). However, Takami does not teach or suggest that the solvent may comprise VEC, or that the solvent must contain both VEC and VC in a concentration of 0.5 to 20 vol % in addition to a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester having no carbon-carbon unsaturated double bond (C) as claimed. Accordingly, even based on the proposed combination of the '473 patent and Takami, there would have been no motivation to utilize a

combination of components (A), (C), and 0.5 to 20 vol % VC and VEC as claimed. Further, Applicants have demonstrated that the use of 0.5 to 20 vol % of the combination of VC and VEC results in a battery with a property which would not have been expected based on the proposed combination of the '473 patent and Takami: inclusion of this concentration of VEC and VC dramatically reduces the amount of gas generated after cycles relative to the amount generated when a greater or lesser amount of the VC/VEC combination is included as component (B).

More specifically, as set forth in the enclosed Fourth Declaration of Atsushi Ueda Under 37 C.F.R. 1.132 ("Fourth Ueda Declaration") seven different batteries were prepared from solvent mixtures each containing a cyclic carboxylic acid ester ((A), here, GBL); a cyclic carbonic acid ester ((C), here, EC); and a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond (B), a combination of VC and VEC in a 1:1 ratio. The relative concentrations of (B) in the solvent mixtures were varied from 0.1 to 30 vol %, as shown in Table 1 of the Fourth Ueda Declaration. It can be seen from the data provided in Table 2 of the Fourth Ueda Declaration that surprisingly and unexpectedly, the inclusion of 0.5 to 20 vol % VC and VEC in a solvent mixture containing GBL and EC (batteries 2-6) significantly decreased the amount of gas generated after cycles compared with batteries containing less than 0.5% VC/VEC (battery 1) or more than 20 vol % VC/VEC (battery 7). The decreases which were observed are dramatic: a reduction of 27% to 57% relative to battery 1 (4.9 ml for battery 1 and 2.1-3.6 ml for batteries 2-6) and 25% to 56% relative to battery 7 (4.8 ml for battery 7).

In sum, it would not have been expected based on the '473 patent, Takami, or their proposed combination that the inclusion of 0.5 to 20 vol % VC and VEC in a solvent mixture containing components (A) and (C) would significantly decrease the amount of gas generated after cycles. Takami does not teach or suggest VEC, and neither Takami nor the '473 patent teaches or suggests the combination of VC and VEC. Thus, the criticality of the concentration of the VC/VEC component would not be expected. Accordingly, the results exhibited by the present invention would overcome any *prima facie* case of obviousness were one to be established, and the battery of the present invention comprising a non-aqueous solvent composed of a cyclic carboxylic acid ester (A), a cyclic carbonic acid ester (B) containing 0.5 to 20 vol% VC and VEC, and a cyclic carbonic acid ester (C) is non-obvious over the proposed combination of the '473 patent and Takami. Reconsideration and withdrawal of the obviousness-type double

patenting rejection based on the '473 patent in view of Takami are respectfully requested.

Rejection Under § 103(a) Based on Takami in view of WO '228 and JP '852

Regarding claims 1-9, the Examiner argues that Takami discloses the claimed non-aqueous electrolyte secondary battery, including the use of a non-aqueous electrolyte containing cyclic carbonates, such as EC, PC, or VC, together with gamma-butyrolactone, and further to be desirable to produce a mixed solvent. Various preferred combinations of non-aqueous solvents are allegedly taught, particularly in paragraphs [0061], [0174] – [0176], [0057] – [0059], and [0272] – [0275]. The Examiner also argues that Takami discloses the claimed positive and negative electrode materials and the employment of lithium salts, such as LiPF₆ and LiBF₄, as well as a solvent containing a benzene-like or derivative compound, as well as a specific solvent mixing solution in Example 41. The Examiner acknowledges that Takami does not disclose the specific use of vinylethylene carbonate (VEC) solvent or its volume percent as claimed.

However, WO '228 allegedly teaches a non-aqueous electrolyte secondary cell having an electrolyte which comprises a cyclic carboxylic acid ester as a non-aqueous solvent and a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond. In particular, the Examiner contends that WO '228 discloses the specific use of vinylethylene carbonate and derivatives thereof, that the content of the cyclic carbonic acid esters is preferably 0.5 to 20 volume %, and that the addition of other esters of not excluded. The Examiner argues that Examples 18-22 show the specific use of VEC added to the cyclic carboxylic acid ester.

Further, JP '852 allegedly discloses a non-aqueous electrolyte secondary battery having a non-aqueous electrolytic solution containing an electrolyte dissolved in a solvent containing at least 20 vol% VC. JP '852 allegedly teaches that VC can be used singly or in combination and that EC and GBL can also be used (paragraphs [0024] – [0025]).

Therefore, the Examiner concludes that it would have been obvious to one having ordinary skill in the art at the time of the invention to use the vinylethylene carbonate solvent of WO '228 in the battery of Takami since WO '228 allegedly discloses that such solvent addition provides a non-aqueous electrolyte secondary cell exhibiting a high electrical conductivity under low temperature circumstances and also suppresses reduction decomposition. The Examiner

contends that since Takami teaches the use of cyclic carbonic acid esters having at least one carbon-carbon unsaturated bond in a combination of mixed solvents, a cyclic carbonic acid ester can be interchangeably used to substitute another cyclic carbonic acid ester because they are deemed to be functionally and chemically equivalent. The Examiner also concludes that it would have been obvious to combine the VC of JP '852 with other solvents to obtain a secondary battery exhibiting high energy density and good cycle characteristics.

Finally, the Examiner again contends that Applicants' previous arguments that the prior art does not demonstrate the unexpected results exhibited by Applicants' invention, specifically for the inclusion of VC and VEC, are not persuasive. Specifically, the Examiner argues that the data which were presented are not commensurate with the claims, noting for example that the gas generating characteristics of the battery have only been shown when the combined volume % of component (B) is 3 vol % and have not been shown for the entire claimed concentration range of 0.5 to 20 vol%. Applicants respectfully traverse this rejection as follows.

In contrast with the present invention, while Takami teaches a mixed non-aqueous solvent, Takami is completely silent as to VEC. Takami thus does not teach or suggest that the solvent may comprise VEC, or that the solvent must contain 0.5 to 20 vol % VEC and VC in addition to a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester having no carbon-carbon unsaturated bond (C) as claimed. WO '228 and JP '852 also do not teach or suggest the combination of VC and VEC and none of the Examples of WO '228 contain such a combination of solvents.

In fact, JP '852 teaches (see abstract) that the ratio of VEC in the non-aqueous solvent is 20 to 60 vol %. JP '852 thus teaches away from the claimed solvent which contains a total of 0.5 to 20 vol % of VC and VEC.

Accordingly, it would not have been expected based on the proposed combination of Takami, WO '228, and JP '852 that the inclusion of 0.5 to 20 vol % VC and VEC in the non-aqueous electrolyte of a secondary battery, as in the present invention, results in dramatic decreases in the amounts of gas generated after cycles compared to batteries in which the non-aqueous solvent contains only VC or VEC or a lesser or greater amount of VC/VEC. Such dramatic decreases have been explained in the Third and Fourth Ueda Declarations and

summarized above or on the record.

Accordingly, these unexpected results would overcome any case of *prima facie* obviousness, were one to be established, and reconsideration and withdrawal of the § 103(a) rejection based on Takami in view of WO '228 and JP '852 are respectfully requested.

Rejection Under § 103(a) Based on Takami in view of WO '228, JP '852 and EP '510

Regarding claims 10-11, the Examiner acknowledges that Takami, WO '228, and JP '852 do not teach that the solvent comprises a glime. However, EP '510 allegedly discloses a non-aqueous electrolyte system consisting of a solvent mixture containing ethylene carbonate, γ -valerolactone and optionally containing one or more additional solvents selected from other organic carbonates such as glymes. The Examiner contends that EP '510 teaches that a mixture of solvents comprising glyme can be used in the electrolyte systems for batteries as they can be applied in a broad voltage range, have a conductivity higher than conventional conductivities at room temperature, and show a high stability against reduction. Therefore, the Examiner concludes that it would have been obvious to one skilled in the art at the time of the invention to use the solvent comprising a glime of EP '510 in the solvent mixture of Takami/WO '228/JP '852. Applicants respectfully traverse this rejection as follows.

As previously explained and demonstrated, Takami, JP '852 and WO '228 do not teach or suggest utilization of a combination of VC and VEC in the non-aqueous electrolyte secondary battery, and Applicants' invention demonstrates unexpected results which would overcome any *prima facie* case of obviousness were one to be established. Specifically, it would not be expected that inclusion of 0.5 to 20 vol % VC and VEC in a non-aqueous solvent containing a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C) would dramatically decrease the amount of gas generated after cycles of the resulting battery, as exemplified by the present invention. EP '510 also does not suggest these results since EP '510 does not teach VC or VEC, nor a solvent which contains, in addition to 0.5 to 20 vol % VC and VEC, a category (A) and a category (C) component. Accordingly, reconsideration and withdrawal of the § 103(a) rejection based on Takami in view of WO '228, JP '852 and EP '510 are respectfully requested.

Rejection Under § 103(a) Based on Takami in view of WO '228, JP '852 and in view of Kida

Regarding claim 14, the Examiner acknowledges that even the proposed combination of Takami, JP '852 and WO '228 does not teach or suggest the claimed molar ratio of both lithium salts. However, Kida allegedly discloses a lithium secondary battery for which it has been reported that the charge/discharge cycle performance can be improved by using both LiPF₆ and LiBF₄ as the electrolytic salts of the non-aqueous electrolyte. Kida allegedly also discloses a specific example using a mixture of LiPF₆ and LiBF₄ in a molar ratio of 4:1. Accordingly, the Examiner contends that it would have been obvious to one having ordinary skill in the art at the time of the invention to use the specific molar ratio of both lithium salts, as taught by Kida, in the electrolyte-solvent mixture of Takami/WO '228/JP '852 to improve the charge/discharge cycle performance and the capacity retention ratio. Applicants respectfully traverse this rejection as follows.

As previously explained, even if the proposed combination of Takami, WO '228, and JP '852 were valid, Applicants' invention demonstrates unexpected results which would overcome any *prima facie* case of obviousness were one to be established. Namely, it would not be expected based on the proposed combination of Takami, JP '852 and WO '228 that inclusion of 0.5 to 20 vol % VC and VEC in a non-aqueous solvent containing a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C) would dramatically decrease the amount of gas generated after cycles. Kida also does not suggest these results, since Kida does not teach or suggest the claimed non-aqueous solvent containing a cyclic carbonic acid ester which contains VC and VEC at any concentration. In particular, Kida teaches in paragraph [0016] that exemplary non-aqueous solvents include ethylene carbonate, propylene carbonate, and gamma-butyrolactone, but are preferably mixed solvents including diethyl ether to give good charge/discharge cycle performance. However, Kida does not teach or suggest VEC, nor a solvent which contains, in addition to 0.5 to 20 vol % VC and VEC, a category (A) and a category (C) component. Accordingly, in view of the unexpected results of Applicants' invention, reconsideration and withdrawal of the §103(a) rejection based on Takami in view of WO '228, JP '852 and Kida are respectfully requested.

Rejection Under § 103(a) Based on Takami in view of WO '228, JP '852 and Inoue

Regarding claim 14, the Examiner acknowledges that even the proposed combination of Takami, JP '852 and WO '228 does not teach or suggest the claimed molar ratio of both lithium salts. However, Inoue allegedly discloses a non-aqueous secondary battery in which the electrolyte comprises one or more lithium salts. It is allegedly preferred to use an electrolytic solution comprising LiPF₆ and LiBF₄ incorporated in a mixture of organic solvents. The Examiner argues that Example 1 shows the use of an electrolyte comprising LiPF₆ and LiBF₄ in an amount of 0.9 mol and 0.1 mol per liter, respectively. Accordingly, the Examiner contends that it would have been obvious to one having ordinary skill in the art at the time of the invention to use the specific molar ratio of both lithium salts as taught by Inoue in the electrolyte-solvent mixture of Takami/JP '852/WO '228 to improve the charge/discharge cycle performance and the capacity retention ratio. Applicants respectfully traverse this rejection as follows.

As previously explained, even if the proposed combination of Takami, JP '852 and WO '228 were valid, Applicants' invention demonstrates unexpected results which would overcome any *prima facie* case of obviousness were one to be established. Namely, it would not be expected based on the proposed combination of Takami, JP '852 and WO '228 that inclusion of 0.5 to 20 vol % VC and VEC in a non-aqueous solvent containing a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C) would dramatically increase the initial charge/discharge efficiency. Inoue also does not suggest these results, since Inoue does not teach or suggest the claimed non-aqueous solvent containing a cyclic carbonic acid ester which contains VC and VEC. In particular, Inoue teaches in col. 12, lines 12-35 that the electrolyte contains at least one aprotic organic solvent such as PC, EC, butylene carbonate, etc., and preferably ethylene carbonate and/or diethyl carbonate. However, Inoue does not teach or suggest VC or VEC, nor a solvent which contains, in addition to VC and VEC, 0.5 to 20 vol% of a category (A) and a category (C) component. Accordingly, in view of the unexpected results of Applicants' invention, reconsideration and withdrawal of the §103(a) rejection based on Takami in view of WO '228, JP '852 and Inoue are respectfully requested.

Rejection Under §103(a) Based on Takami in view of WO '228 and JP '852 and in view of Hamamoto

Regarding claim 15, the Examiner acknowledges that the proposed combination of Takami, JP '852 and WO '228 does not disclose that the solvent comprises a derivative of benzene. However, Hamamoto allegedly teaches in the abstract that a non-aqueous electrolytic solution which may be used for a lithium secondary battery employs a non-aqueous electrolytic solution which comprises a non-aqueous solvent and an electrolyte which contains a biphenyl derivative. In view of these teachings, the Examiner concludes that it would have been obvious to one having ordinary skill in the art at the time of the invention to use the solvent of Hamamoto comprising a derivative of benzene in the solvent mixture of Takami, JP '852 and WO '228, as Hamamoto teaches that by using the benzene derivative as a solvent, a non-aqueous electrolytic solution which is favorably employable for a lithium secondary battery and which shows high battery performance, such as high electric capacity and high cycling performance, under maximum operation voltage condition or elevated temperature is obtained. Applicants respectfully traverse this rejection as follows.

As previously explained, even if the proposed combination of Takami, JP '852 and WO '228 were valid, Applicants' invention demonstrates unexpected results which would overcome any *prima facie* case of obviousness were one to be established. Hamamoto also does not suggest these results, since Hamamoto does not teach or suggest the claimed non-aqueous solvent containing a cyclic carbonic acid ester which contains VC and VEC. In particular, Hamamoto in paragraph [0018] teaches that the non-aqueous solvent preferably comprises a combination of a cyclic carbonate and a linear chain carbonate, a high dielectric constant solvent such as ethylene carbonate, propylene carbonate, or butylene carbonate, and a low viscosity solvent, such as γ -butyrolactone or dimethyl carbonate, for example. However, Hamamoto does not teach or suggest VC or VEC, nor a solvent which contains, in addition to 0.5 to 20 vol % VC and VEC, a category (A) and a category (C) component. Accordingly, in view of the unexpected results of Applicants' invention, reconsideration and withdrawal of the §103(a) rejection based on Takami in view of WO '228, JP '852 and Hamamoto are respectfully requested.

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In view of the preceding remarks and Fourth Ueda Declaration, Applicants respectfully submit that all of the pending claims are patentably distinct over the prior art of record and in condition for allowance. A Notice of Allowance is respectfully requested.

Respectfully submitted,

ATSUSHI UEDA et al.

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(Date)

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Encl: Fourth Declaration of Atsushi Ueda Under 37 C.F.R. § 1.132